

# Temporal Coherence of Semiconductor Excitons across Bose-Einstein Condensation

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Very recently, we have shown experimentally that Bose-Einstein condensation leads to a mostly dark gas of excitons in GaAs heterostructures. A small fraction of optically active states nevertheless contributes to the condensate so that a weak photoluminescence manifests the excitons non-classical spatial coherence. In this Letter, we show that this photoluminescence experiences a threshold increase of temporal coherence at the critical temperature for excitonic condensation.

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In semiconductor physics, Bose-Einstein condensation has received a large attention. Indeed, semiconductor excitons are light-mass composite bosons which were predicted already in the 1960s to undergo Bose-Einstein condensation [1–3]. Demonstrating the quantum phase transition initially held good premises, because it is expected under reasonable experimental conditions, precisely below a few Kelvin in widely studied GaAs heterostructures. Unfortunately, first hope underestimated the role of the exciton electronic structure: the spin degree of freedom of Coulomb bound electrons and holes leads to optically active and inactive excitonic states, which greatly complicates the detection of a condensate.

For excitonic condensation, the key role of dark states was pointed out recently by Combescot and co-workers [4]. They stressed that dark excitons always lie at the lowest energy, because they do not experience repulsive interband Coulomb scatterings mediated by virtual photons. Consequently, Bose-Einstein condensation leads to a macroscopic population of dark excitons somewhat hiding the buildup of the condensate. In fact, quantum signatures can only be detected directly above a density threshold, when fermion exchanges between excitons introduce a coherent small fraction of bright excitons to the dark condensate [5]. The latter then becomes “grey” and is possibly studied through its weak coherent photoluminescence [6].

In GaAs double quantum wells, where two-dimensional indirect excitons can be engineered by spatially separating electrons and holes [7], we have recently observed unambiguous signatures for the predicted grey condensate of excitons. Using an electrostatic dipole trap [8–12], we have first reported the buildup of a non-classical distribution, with about 80% of excitons occupying dark states at a bath temperature  $T_b = 330$  mK [13]. This distribution is non-classical because dark states are only separated from bright ones by a few  $\mu\text{eV}$  in double quantum wells [14], that is, ten times less than the thermal energy. More recently, we have demonstrated in this dark and dilute regime that indirect excitons can realise a complex superfluid state distributed between the four lowest energy bright and dark states [15]. This behaviour

is found below a critical temperature of about 1 Kelvin when indirect excitons emit a weak photoluminescence with macroscopic spatial coherence.

The photoluminescence radiated by a grey condensate then reveals its electronic properties. In particular, we expect a spectral narrowing of the photoluminescence across the quantum phase transition because the emergence of macroscopic spatial coherence implies exciton condensation in momentum-space. This behaviour is revealed in this Letter where we confine  $\sim 10^4$  indirect excitons in a  $10\text{ }\mu\text{m}$  wide trap, as in Ref. [15]. Using time and spatially resolved interferometry, we show that the coherence time of the photoluminescence radiated from the trap is controlled by the bath temperature: above a critical temperature  $T_c \sim 1$  K the coherence time is about 2 ps and suddenly increases to  $\sim 4$  ps below threshold. This transition coincides with the previously found threshold for macroscopic spatial coherence in the photoluminescence signal [15]. We thus conclude that these coincident thresholds constitute the fingerprints for Bose-Einstein condensation of excitons in GaAs heterostructures.

Beyond studies of collective quantum phenomena, our work also opens a route to quantify exciton-exciton interactions through the photoluminescence spectrum. For that purpose, one needs ideally to probe a gas at thermal equilibrium and free from inhomogeneous broadening. This regime is accessible to indirect excitons. We note that Zimmerman has predicted a Lorentzian photoluminescence with  $\approx 200\text{ }\mu\text{eV}$  spectral width in experimentally studied conditions, e.g. at a few Kelvin [16]. However, most experiments have reported values almost ten times larger [11, 17–20] in often asymmetric photoluminescence spectra. Instead, here we study a homogeneously broadened gas, with a Lorentzian photoluminescence  $\sim 300\text{ }\mu\text{eV}$  spectrally wide. In this regime, theoretical treatments of exciton-exciton interactions [16, 21–24] are easily compared to experiments.

As shown in Fig.1.b, here we study indirect excitons (IXs) made of electrons and holes confined in two different 8 nm GaAs quantum wells separated by a 4 nm Al-GaAs barrier. Thus, the overlap between electrons and holes wavefunctions is decreased, so that indirect exci-

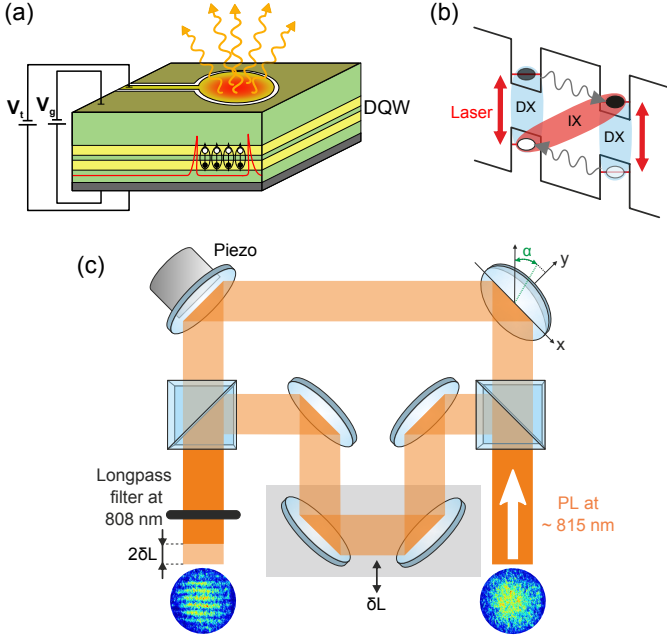


FIG. 1: (a) Our device consists of a double quantum well (DQW) embedded in a field-effect device made of two electrodes: A central  $10\ \mu\text{m}$  disk, the trap region, separated by a  $200\ \text{nm}$  gap from the outer guard electrode. We apply the same bias to both electrodes,  $V_c = V_g = -4.7\ \text{V}$ , to imprint a hollow trap illustrated by the red line confining indirect excitons. (b) Indirect excitons (IX) are formed after relaxation of electrons and holes injected in both quantum wells, by exciting resonantly the direct-exciton (DX) absorption. (c) The photoluminescence emitted by  $\sim 10^4$  excitons confined in the trap is analysed by a Mach-Zehnder interferometer. The two arms have vanishing lateral shift and a variable longitudinal path length difference  $\delta L$ . At the output, horizontal interference fringes are realised by tilting one of the mirrors at an angle  $\alpha \sim 28^\circ$ .

tons acquire a long optical lifetime ( $\sim 70\ \text{ns}$ ) and can reach thermal equilibrium after optical injection [13]. As in previous works, we use a  $10\ \mu\text{m}$  electrostatic trap to spatially confine indirect excitons, the trapping potential being controlled by two metallic electrodes deposited at the surface of the field-effect device embedding the double quantum well. We apply the same potential on both electrodes ( $V_t = V_g = -4.7\ \text{V}$ ), so that the  $200\ \text{nm}$  gap between them creates a rectification of potential leading to an electrostatic barrier, i.e. to a hollow-trap [15, 25].

In our studies, indirect excitons are optically injected using a  $100\ \text{ns}$  long laser pulse ( $P_{\text{exc}} = 700\ \text{nW}$ ) covering most of the trap area. The laser is set resonant with the direct excitons absorption for a minimum laser-induced heating. We then study the photoluminescence (PL) due to the radiative recombination of bright IXs. The photoluminescence is collected by the same objective as the one used for laser excitation, yielding a  $\sim 1\ \mu\text{m}$  spatial resolution. As in previous works, we emphasise the long delays regime after extinction of the loading laser pulse: the photoluminescence is studied in a  $5\ \text{ns}$

long time interval, set between  $180$  to  $210\ \text{ns}$  after the end of the laser light, the sequence being repeated at  $1.5\ \text{MHz}$ . We concentrate on an experimental realisation where  $\approx 10^4$  excitons are confined in the trap,  $\sim 80\%$  of them contributing to the grey condensate formed at a bath temperature  $T_b = 330\ \text{mK}$  [13, 15].

To scrutinise the impact of quantum condensation onto the excitons temporal coherence, we magnified the real image of the photoluminescence and sent it into a Mach-Zehnder interferometer, as sketched in Fig.1.c. The PL is divided equally between the fixed arm and the mobile arm. At the output of the interferometer, the two beams are superposed, so that there is no spatial displacement. The temporal coherence of the PL is inferred by varying the longitudinal path length difference, i.e. by translating two mirrors of the mobile arm by  $\delta L$ . The two outputs are thus temporally shifted by  $\delta t = 2\delta L/c$  where  $c$  is the speed of light in vacuum. In our experiments, the path length difference is actively stabilised: we typically reach a  $\delta L$  deviation below  $20\text{-}30\ \text{nm}$  with day-long stability.

The contrast of the interference pattern provides a direct measurement for the coherence time  $\tau_c$  of the optically active excitons. Indeed, the interference visibility is given by the modulus of the first-order correlation function  $|g^{(1)}(\delta t)| \propto |\langle \psi_0^*(\mathbf{r}, t) \psi_0(\mathbf{r}, t + \delta t) \rangle_t|$ . Here,  $\psi_0(\mathbf{r}, t)$  is the photoluminescence field which reflects the wave function of the lowest energy bright excitons, while  $\langle \dots \rangle_t$  denotes the time averaging and  $\mathbf{r} = (x, y)$  reads the coordinate in the plane of the quantum well. For a homogeneously broadened gas, the  $g^{(1)}$ -function is exponentially decaying with  $\delta t$ , at a rate  $1/\tau_c$ . In a thermal regime above a few Kelvin, it is expected that  $\tau_c$  is controlled by exciton-phonon interactions [26]; however, below a few Kelvin, we expect a dramatic change because these interactions become inefficient.

Unlike studies realised with atomic gases [27], our experiments require an accumulation over several millions of single-shot realisations. In this situation, the stability of the electrostatic trap confining indirect excitons, bound to  $\sim 500\ \mu\text{eV}$  [13], is a severe experimental limitation. Indeed, we measure very different PL spectra in successive  $15\text{-second}$  long acquisitions. This is illustrated in Fig.2.a showing a spectrum made of a central Lorentzian line, with a width limited by the spectrometer resolution, together with a low amplitude and broader background. By contrast, Fig.2.b shows a broad spectrum, about  $1\ \text{meV}$  wide with no recognisable shape. Spectral fluctuations obviously have a strong impact on the interference pattern of the PL, even if this alternative allows reducing the acquisition time. As shown in Fig.2.c-d, we can either observe clear fringes all across the center of the trap, or very blurred patterns where fringes are hardly distinguished. This limitation motivated studies of an ensemble of interference patterns from which we extract the contrast by averaging statistically the best  $10\%$  of  $6\text{-second}$  expositions, to enforce a signal to noise ratio of about  $8$  on each exposition.

Figure 3 quantifies the variation of the interference

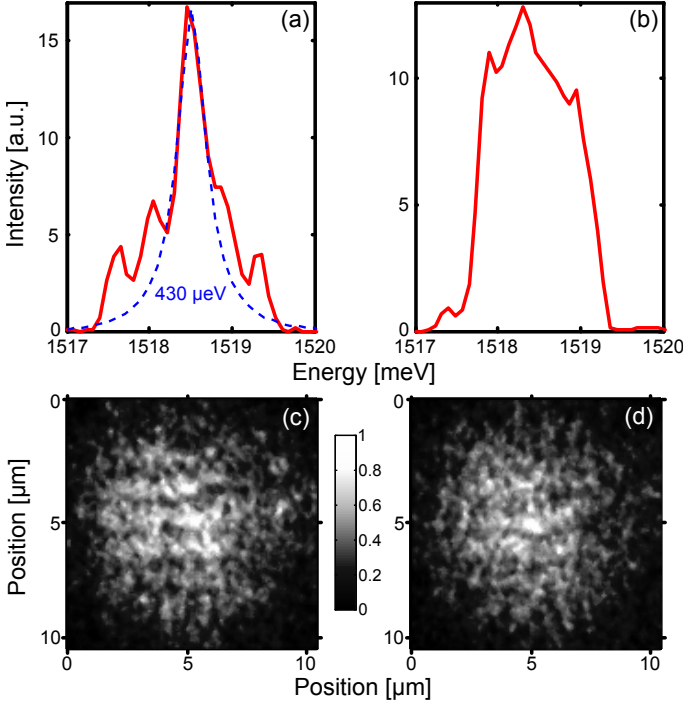


FIG. 2: (a-b) Spectrally resolved PL at  $T_b = 330$  mK. Measurements are realised during a 15 second long acquisition. In (a) the central sharp line is fitted with a Lorentzian shown by the dashed blue line (FWHM =  $430 \mu\text{eV}$ ). (c-d) Interference patterns measured in the same experimental conditions as (a-b). These measurements are realised during a 12-second long acquisition, the path length difference being set to  $\delta t = 1.8$  ps.

contrast along the center of the trap, as a function of the time delay  $\delta t$  introduced between the two arms of the interferometer. At every bath temperature probed in these measurements,  $T_b = 0.33$  to  $2.36$  K, the visibility follows a mono-exponential decay  $e^{-|\delta t|/\tau_c}$ . According to the Wiener-Kintchine theorem [28], the photoluminescence spectrum then consists of a single Lorentzian in these measurements. This shows that we study a homogeneously broadened gas which constitutes a major improvement regarding previous works [18, 20]. To the best of our knowledge, such a single Lorentzian PL profile had never been observed for a gas of indirect excitons.

As expected, we note in Fig.3 that the decay time of the  $g^{(1)}$ -function largely increases when the bath temperature is lowered from  $2.36$  to  $0.33$  K. To quantify this behaviour, we fix  $\delta t = 1.8$  ps thus optimising the sensitivity of our measurements. Figure 4.a shows that two regimes emerge: For  $T_b \geq 1.3$  K,  $|g^{(1)}|$  varies weakly, it is about 22%, corresponding to a coherence time  $\tau_c \sim 2$  ps. By contrast, for  $T_b \leq 1.3$  K, the interference contrast increases steplike to 32% at  $T_b = 330$  mK, manifesting that  $\tau_c$  is doubled at sub-Kelvin bath temperatures, as shown in Fig.4.b.

Figure 4 reveals that optically active bright excitons suddenly gain temporal coherence in the trap below a

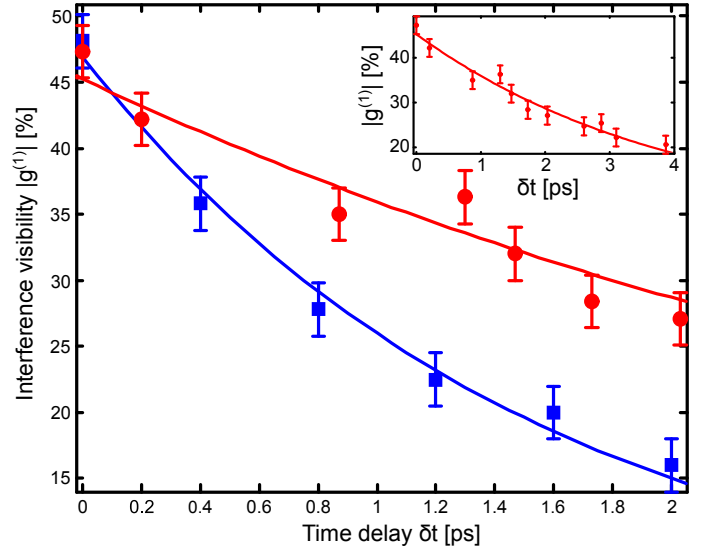


FIG. 3: Interference visibility measured along the center of the trap as a function of the time delay introduced in the interferometer. Measurements taken between  $T_b = 330$  mK (red circles) and  $2.36$  K (blue squares) all follow single exponential decays (solid lines). The insert displays the data at  $T_b = 330$  mK with an extended abscissa, fitted by a single exponential with a time decay of 4 ps. Interference contrasts are extracted from a sample of 100 acquisitions, for every data point.

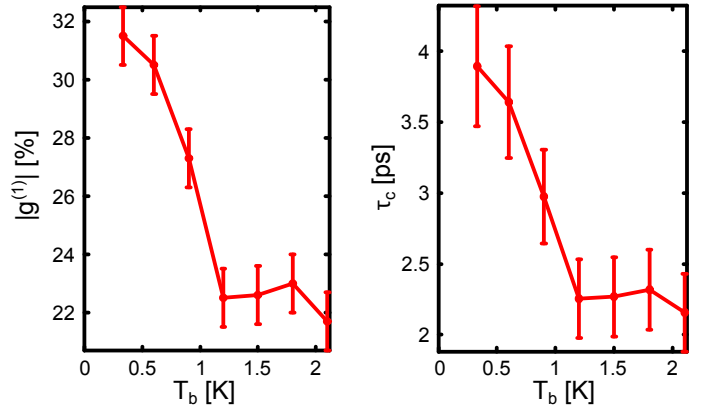


FIG. 4: (a) Interference contrast  $|g^{(1)}|$  measured at the centre trap as a function of  $T_b$ . (b) Photoluminescence coherence time  $\tau_c$  as a function of the bath temperature  $T_b$ . The time delay is fixed at  $\delta t = 1.8$  ps in these measurements.

critical temperature of about 1 K. We have very recently shown that they concomitantly acquire macroscopic spatial coherence which signals the formation of a four-component grey condensate of indirect excitons [15]. As previously mentioned, we expected this coincident buildup of temporal and spatial coherences: The spatial coherence signals that the condensate wave-function is essentially limited by the size of the trap. On the other hand, below the critical temperature  $T_c \sim 1$  K, the fraction of excitons in the condensate grows rapidly, which

necessarily implies an increase of temporal coherence. Indeed, exciton-exciton interactions tend to become more important for the relative occupations of optically bright and dark states than for the dephasing of the collective wave-function.

At our lowest bath temperature,  $\tau_c$  remains about 4 orders of magnitude lower than the excitons radiative lifetime ( $\sim 30$  ns [13]). This difference raises intriguing questions by signalling a high scattering rate out of the condensate, towards higher energy states. The resulting non-condensed part amounts to less than 20% of the total exciton population at  $T_b = 330$  mK, as shown by spatial interferometry [15]. Using temporal interferometry, here we show that excited states are indeed efficiently populated. In future work, we aim at raising quantitative conclusions regarding the processes governing  $\tau_c$ . For that purpose, technical limitations will have to be surpassed since electrostatic instabilities put strong limits

on the acquisition time, i.e. on the number of single-shot realisations we can accumulate.

To conclude, we have shown that Bose-Einstein condensation leads to a two-fold and steplike increase of the temporal coherence of excitons trapped in GaAs quantum wells. The buildup of temporal coherence coincides with the buildup of the non-classical spatial coherence [15]. We reveal this behaviour by probing a homogeneously broadened gas of dipolar excitons in a trap at fixed density and controlled temperature.

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